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COMMITTEE ON THE SANITARY ENGINEERING ASPECTS OF NUCLEAR ENERGY - A PROGRESS REPORT

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Oak Ridge, Tennessee

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COMMITTEE ON THE SANITARY ENGINEERING ASPECTS OF NUCLEAR ENERGY

A PROGRESS REPORT*

OCTOBER 1954

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* Presented at the Annual Meeting, American Society of Civil Engineers, New York, New York, October 19, 1954.

The Committee on the Sanitary Engineering Aspects of Nuclear Energy was activated at the Chicago, Illinois meeting of the Executive Committee, Sanitary Engineering Division, ASCE, February 21-22, 1953. The purposes of this Committee were outlined as follows: to advance the body of knowledge involving sanitary engineering and radiological health, to stimulate interest and activity in this field by sanitary engineers, to increase the sanitary engineers' know-how in this field, to recommend committees for special task investigations and reports, and to recommend publications of manuals and papers on subjects in this field.

The following individuals were designated to membership on the Committee: Mr. S. T. Barker, Assistant Director, Bureau of Environmental Sanitation, New York State Department of Health, Albany, New York; Mr. Arthur E. Gorman, Sanitary Engineer, Division of Engineering, U.S. Atomic Energy Commission, Washington, D. C.; Dr. Warren J. Kaufman, Assistant Professor of Sanitary Engineering, University of California, Berkeley, California; Mr. James G. Terrill, Jr., Chief, Radiological

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Health Program, Public Health Service, Department of Health, Education, and Welfare, Washington, D.C.; and Chairman, Dr. Conrad P. Straub, Senior Sanitary Engineer, Robert A. Taft Sanitary Engineering Center, Public Health Service, Department of Health, Education, and Welfare, on assignment to the Oak Ridge National Laboratory, Oak Ridge, Tennessee.) Designated contact member was Mr. Roy J. Morton, Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee.

This report of the Committee is divided into four parts, the first indicates very briefly what the Committee's activities have been since its inception, the second includes a statement of the efficiency of conventional water and sewage treatment processes for the removal of radioactive materials, the third covers the radioassay of liquids, and the fourth considers future activities of the Committee.

ACTIVITIES 1953-1954

The major effort of the Committee has been expended in the promotion of a column in CIVIL ENGINEERING entitled "Nuclear Notes." The purpose of this column is to acquaint the practicing engineer with some fundamental information to better understand and appreciate the value of nuclear energy in the solution of his engineering problems. The column appeared in the May, 1954 issue of CIVIL ENGINEERING and has appeared monthly since that time. Following a brief review of possible applications of nuclear energy to civil engineering problems, the topics discussed have included the structure of the atom, nuclear particles and rays, natural radioactivity, nuclear reactions, and

neutron reactions. With the appearance of the first column several requests have been received for additional information relating to specific applications of nuclear energy to problems encountered by the practicing engineer, as well as requests for further information to clarify some of the points discussed. Such requests are welcomed by the Committee, and the Committee is interested in learning about any applications of nuclear energy or the use of radioactive materials in the solution of engineering problems. We want the members to feel that this is their column and we invite comments, suggestions, criticisms, etc.

The Committee assisted in the preparation of comments on the proposed amendment to the U.S. Atomic Energy Act. A statement of testimony representing the position of the ASCE with respect to the amendment was based on these comments.

REMOVAL OF RADIOACTIVE MATERIALS BY CONVENTIONAL WATER AND SEWAGE TREATMENT PROCESSES

The second and major portion of this report is concerned with an evaluation of conventional water and sewage treatment processes for the removal of radioactive materials from liquids. There is considerable misunderstanding on the part of the engineering profession on the value of water and sewage treatment processes for the removal of radioactive materials. Originally, it was thought that conventional coagulation, settling, filtration, and some of the other water treatment processes and biological sewage treatment methods would be especially effective

in the removal of specific radioactive materials. Experience in the laboratory, however, has not borne out this optimism.

In the first place, it must be remembered that radioactive and stable isotopes of an element behave in the same way during chemical and biological treatment. Thus, if the conventional process is not designed to remove a specific element, there is no reason to believe that the process will remove the specific radioisotope. Radioactive calcium, barium, or strontium, for example, will be significantly removed by softening, since the purpose of softening is to remove calcium. The second consideration is that because of the extremely minute concentrations of radioactive materials in waters or waste solutions presently discharged to the environment, adsorption on mineral or biological substances may be an important factor in their removal, particularly for those radioisotopes of relatively scarce elements of high valence, such as yttrium, zirconium, niobium, and the rare earths. By such methods, it may be possible to remove some contaminants that are normally foreign to natural waters. As pointed out later, however, even this mechanism of removal has not shown too much promise.

Maximum Permissible Concentrations of Radioisotopes

Before discussing the efficiency of removal processes, it is well to call attention to the maximum permissible concentrations (MPC) that have been established for specific radioactive materials in water and air, based on maximum permissible body burdens. These values, which have been published in NBS Handbook 52[1], are based on the assumption

that a person will use, in the case of water for example, a source contaminated by the radioactive materials for a period of 70 years. In other words, this will be his sole source of supply, and if the limiting concentrations are not exceeded, there is little likelihood that damage will result from such ingestion. If one were to consume the waters for considerably shorter periods of time, higher values could be permitted. Such values have been released by the Atomic Energy Commission[2] and cover safe concentrations in water immediately after a nuclear detonation.

A study of the values in Handbook 52 and reactor fission yield data shows the most hazardous radioisotopes in fission products are Sr^{89}, Sr^{90}-Y^{90}, Y^{91}, Zr^{95}-Nb^{95}, Ru^{106}-Rh^{106}, I^{131}, Cs^{137}-Ba^{137}, Ba^{140}-La^{140}, and Ce^{144}-Pr^{144}. These and P^{32} (because of its wide use as a tracer) will be of most interest to the water and sewage works operator. With few exceptions, it may be stated that on the basis of present off-site usage of radioactive materials in this country, there is no immediate problem of significant radioactive contamination of our water courses.

Conventional water and sewage treatment processes will be useful only in those cases where the levels of radioactive materials are relatively low, certainly in the microcurie per liter (μc/l) range. If there is continuous discharge of Sr^{89} into a source of water supply, for example, the highest permissible discharge, based on an MPC value of 7×10^{-5} μc/ml for this radioisotope and a removal efficiency of 75 per cent through a softening plant, will be about 1×10^{-4} μc/ml or

3.4×10^{-9} ppm, which is an extremely low concentration.

Before a method of treatment for the removal of radioactive materials can be suggested or provided, the sanitary engineer, consulting engineer, or water works operator must know several things: 1) the concentration of radioactive materials in the liquid to be treated, 2) the composition of radioactive materials in the source, 3) the MPC of the individual components comprising the radioactive waste mixture, and 4) the efficiency of the particular process that he intends to employ for the removal of the various radioelements. To obtain information on item 1 he must know what radioisotopes are used on the watershed, in what amounts, and in what concentrations they are discharged. If only a single radioisotope is found, item 2 presents no problem. Where several radioisotopes are used, however, radiochemical analyses must be made, and these become less accurate as the number of radioisotopes in the mixture increases. Information on item 3 will be found in Handbook 52. Item 4 will be considered subsequently. If radioisotopes are encountered for which no information relative to the efficiency of removal afforded by the various treatment processes has been published, it may be possible for the water or sewage works operator to carry out such studies to determine how these radioactive materials will behave in passing through his plant.*

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* The mode of analysis outlined in this paragraph would give only an order-of-magnitude estimate. There are too many uncertainties in making such studies and analyses, since tests would have to be made of specific materials. Furthermore, very few water plant operators could make such tests.

Effectiveness of Water Treatment Processes

With the exception of most cations of valence 3, 4, or 5, including the rare earths group, coagulation and settling has not been very effective for the removal of radioactive materials from water[3]. Phosphorus has been removed from water and waste solutions[3][4], but the preparation of the influent water is of importance here[4]. Considerable care must be given to the coagulation step. Iodine, which has found extensive use in medical therapy and research, cannot be removed by coagulation with alum or iron[3][4][5]. However, the addition of small amounts of activated carbon[3], copper sulfate[3], or silver nitrate[3][5], has materially increased the removal of this anion from water.

Sand filters, except for removal by simple straining or adsorption on biological life contained on the "Schmutzdecke", have not been effective[6][7]. Their major function is to remove the radioactivity previously incorporated in floc particles.

Softening with lime and soda ash has been studied[8][9][10][11], and under proper conditions, effective removals of strontium, barium, cadmium, yttrium, scandium, and zirconium-niobium have been obtained[8]. For most satisfactory removals excesses of both lime and soda ash are required[9]. Some recent studies have indicated that the strontium present is removed by coprecipitation[9][10] with calcium carbonate by the mechanism of mixed crystal formation. Recently, data on the removal of stable strontium by municipal water treatment processes has been reported[12].

Removal of radioactive materials by ion exchange is technically possible[11][13][14], and for most effective removal mixed bed or cation and anion exchange beds in series is recommended[13][14].

Several modifications of existing processes have been studied to obtain better removal of radioactive materials. The most promising has been the use of phosphate coagulation[15][16]. Relatively high removals have been obtained for many radioisotopes of interest[15], including strontium, but in the case of the latter, these removals were obtained under carefully controlled conditions of pH and the calcium hydroxide to trisodium phosphate ratio[15][16]. Other methods suggested include the use of metal powders[17] and added clay materials[3][18].

In the case of mixed fission products, the efficiency of removal is a function of the fission products mixture. Removals reported to date have been in the order of about 70 to 80 per cent for alum and iron coagulation[3]. A series of studies on actual process waste waters at Oak Ridge National Laboratory disclosed that removals up to 90 per cent could be obtained with excess lime-soda ash softening or with phosphate coagulation when coupled with the addition of clay, which is specific for the removal of cesium[19].

In addition to the presence of radioactive materials in the environment by direct discharge into the water carriage system, there is also the possibility of deposition of radioactive materials in water courses as a result of fall-out following weapon's test activities. Where many of the radioactive materials discharged directly into our surface

streams will be in solution, many of these, deposited along with fall-out, may be in the form of insoluble oxides, and removal of these may differ from the removal of soluble ions.

Several studies have been reported recently dealing with the problem of fall-out in particular areas[20][21][22]. At Albany, New York[20], it was reported that about 35 per cent of the activity due to fall-out was removed in passing through the Albany water supply system and treatment plant, which included alum coagulation, settling, and filtration, whereas studies reported by Thomas et al.[21] covering operation of the Lawrence, Massachusetts water treatment plant showed an overall removal of 80 per cent following coagulation, settling, and filtration. Samples analyzed at Cincinnati[22] indicated that from 50 to 100 per cent of the natural radioactive materials in rain water during normal times were soluble, but during the weapon's testing program the soluble fraction of radioactive materials was reduced to approximately 30 per cent.

Effectiveness of Sewage Treatment Processes

The adsorptive properties of aerobic biological floes for the organic components of sewage led early researchers to hold great promise for the application of sewage treatment processes for the removal of radioactive wastes. This expectation has not been entirely substantiated in the research laboratory. The present consensus indicates that the biological concentration of radioisotopes from liquid wastes may have only limited application. Several research groups have reported up to 94 per cent removal of plutonium by the activated sludge and standard rate trickling filter processes[6][23][24]. It has been suggested also

that biological treatment of wastes containing organic sequestering agents may be desirable prior to chemical precipitation of the radioactive contaminants. With these two exceptions, there is a preponderance of evidence that biological flocs and slimes have relatively limited capacity to assimilate mono- and di-valent radioisotopes, particularly those which are soluble and are abundantly present in stable form[25][26][27]. This limitation has led to the conclusion that in situations requiring decontamination factors of 10 (90 per cent removal calculated by dividing influent activity by effluent activity) or more it is unlikely that biological systems will be satisfactory. In some instances where sewage treatment is necessary for the abatement of stream pollution, the conventional process may serve also to reduce the load of radioactive materials in the receiving water.

The uptake of radioactive iodine and phosphorus by sewage treatment processes has been of some interest since these two isotopes are widely employed by medical and research groups and after use are generally discharged to sewers. Several studies of iodine-131 removal by trickling filters have been completed with efficiencies ranging from a few per cent up to 85 per cent[26][29][30]. Similar research with the activated sludge process[25][26] indicates removals may range from one per cent to 97 per cent and are largely dependent upon the concentration of stable iodine-127 present in the waste. As little as 0.1 ppm of stable iodine may reduce removals to 20 per cent or less. Studies of phosphorus-32 uptake by activated sludge further emphasize the importance of carrier influence by isotope dilution[25][26]. The stable phosphorus uptake of a biological

system, together with the phosphorus concentration of the waste, will essentially fix the degree of uptake of radioactive phosphorus. Generally, not more than 20 to 30 per cent removal of radiophosphorus will be obtained from domestic sewage, either by the activated sludge or trickling filter processes. Somewhat higher removals may be expected in oxidation ponds operating with short detention periods. It should be stressed that radioactive iodine and phosphorus are not considered to be potentially serious radiological health hazards at the present or probable future levels of use.

Considerable interest has been shown in the treatment of contaminated laundry wastes[31][32]. Up to 90 per cent removal of mixed fission products was obtained with single stage standard rate trickling filters operated with a recirculation ratio of 3. In numerous studies of the biological uptake of fission products from natural and synthetic sewages, removals have consistently ranged from 70 to 85 per cent[4][25][27][30]. With the activated sludge process, variations in aeration periods and sewage strengths have little influence on the removal of mixed fission products. In this instance it is probable that the fraction of activity absorbed is represented by particular elements that individually are almost completely removed.

The removal of strontium-90 is of particular interest since it is the most hazardous of the fission product group. The presence of calcium and magnesium, as well as stable strontium, has been shown to markedly reduce the radiostrontium uptake. Reported removals range from 11 to 99 per cent. Probably the wide variation is due to variations

in waste composition[6][27][32]. It is likely that where the only mechanism of removal is biological assimilation , efficiencies will not exceed 50 to 75 per cent.

Only limited data are available on the effectiveness of oxidation ponds for the concentration of radioisotopes and these data offer no indication that this process has any particular advantage over those previously discussed. Removals of mixed fission products have been reported ranging from 10 to 80 per cent while that of radioactive strontium did not exceed 20 per cent[33].

It has become apparent that in the treatment of low level radioactive wastes radiation will have no significant influence on the biological flora and fauna. It is also very probable that the levels of radioactivity likely to be encountered at the municipal sewage treatment plant will have no influence on the treatment process. These conclusions stem from the fact that the inefficiency of biological treatment will permit its application for the treatment of only low level wastes if our current standards for maximum permissible concentrations in natural waters are to be met. At these levels the influence of radiation is not significant.

THE RADIOASSAY OF LIQUIDS

One of the most immediate needs of the sanitary engineering profession in the area of nuclear science is a practical and working understanding of methods of detecting radioactivity. In the evaluation of water contamination hazards resulting from the nuclear weapon's

testing program or from the operation of nuclear reactors, it is imperative that reliable measures of radioactivity be made at levels in the order of 10^{-8} μc/ml. The absolute determination of radioactivity at these low concentrations is fraught with error, both statistical and systematic in nature. To correctly assess the radiological health hazards resulting from mixtures of radioisotopes the radiochemical separation and assay of the individual constituents will certainly be desirable.

Although numerous sanitary utilities are now equipped with detection equipment and are currently performing radioassays the data thus obtained may be of limited value and comparability.

Probably the most definitive step in solving this problem has been made by the personnel of the Robert A. Taft Sanitary Engineering Center in the form of a simple procedure for the gross radioassay of water[34] for beta and alpha activity. It is generally agreed that the evaporation technique followed by assaying with an internal proportional counter affords the greatest sensitivity and precision. An index of the radioactivity associated with particulates can be obtained by first filtering the samples to be analyzed through a hydrosol membrane and making a direct count of the filter. The dissolved radioactivity may be assayed by evaporating 200 to 1,000 ml of the filtrate and counting the residue. Corrections must be made for backscattered radiation, for loss of radiation by self-absorption, and for the geometric efficiency of the counting instrument.

The backscatter and self-absorption corrections are dependent

on the method of mounting the sample and on the radioisotopic composition and mineral content of the liquid to be assayed. Since these corrections are difficult to evaluate for a sample of unknown composition and may display considerable variance, it is essential that the method of sample preparation and mounting be carefully standardized. As a possible solution to this problem it has been suggested that the thallium-204* isotope be

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* Thallium-204 is a good choice for a standard sample because it has a reasonably long half-life, 2.7 yrs, it emits no gamma radiation and a single beta particle of 0.78 million electron volts (Mev). This energy (0.78 Mev) is close to the average for a fission product mixture. However, if counting corrections are made to Th^{204}, one might overestimate the activity of a Sr^{89} sample where the beta particle has an energy of 1.5 Mev and underestimate the activity of a S^{35} sample where the beta particle has an energy of 0.166 Mev.

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utilized as a standard and that all corrections be incorporated into a single factor by simply ascertaining the overall counting efficiency in terms of this arbitrary standard(35). Under these circumstances the activity of an unknown source would be reported as the "thallium-204 equivalent activity." If the level of radioactivity were sufficiently great, it would be desirable to perform a radiochemical analysis and to report the concentration of individual isotopes. This latter phase is particularly difficult, but work is currently underway to develop a simple procedure for separating the more hazardous radioisotopes. Thallium-204 standard solutions are available from the National Bureau of Standards.

The literature is replete with basic data concerned with absolute beta counting(36)(37)(38)(39)(40)(41) and describing statistical

considerations[42][43] pertinent to the detection of low levels of activity. It now remains for the sanitary chemists and engineers to adopt a procedure that will enable the making of reliable radioassays at the state health department and major water utility laboratory levels.

FUTURE ACTIVITIES

During the coming year, the Committee will continue the series "Nuclear Notes" in CIVIL ENGINEERING, and will encourage the presentation of papers in the nuclear energy field and publication of these as ASCE Separates.

With the interest in the use of nuclear energy as a source of competitive power, and the future expansion of this phase of nuclear energy, the environmental problems dealing with the handling and disposal of effluvia from these plants will offer considerable challenge and opportunity for the sanitary engineer. He must be prepared to meet this challenge.

Since our knowledge of the nature, composition, and fate of fallout particles is extremely limited, the Committee suggests that greater emphasis be placed on a study of this problem. What happens to this material after it has precipitated from the atmosphere or been carried along with rainfall? How much of the total amount precipitated or deposited is actually carried away as runoff and enters the receiving stream? How much of it will be removed in the water or sewage treatment plant? How much of the material is soluble and how much

insoluble? What is the size of the particulate matter, what radio-isotopes are present, and what is the activity of the particles? How much of the fallout material is taken up by plants which may be consumed by man or animals? Considerable information is needed to answer these and similar questions. Perhaps such data can be obtained during the course of future weapon's testing programs; it will be of great value in Civil Defense and other planning. The Sanitary Engineering Division, ASCE, should encourage such research and should strongly recommend that these studies be made.

It is hoped that the Committee roster will be broadened to include representation from the consulting engineering office, the power interests, the Federal Civil Defense Administration, as well as the agricultural interests.

Respectfully submitted,

Conrad P. Straub, Chairman
Stanley T. Barker
Arthur E. Gorman
Warren J. Kaufman
James G. Terrill, Jr.

10/9/54
revised: 12/20/54.

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